

# Kinetics Of Dissipation And Biodegradation Of Crude Oil In Alaska's Cook Inlet

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## Abstract

*The results of a study to quantitatively define the magnitude of oil pollution problems in Alaska's Cook Inlet are reported. Physical dissipation and biodegradation rates were determined and combined with estimates of hydrocarbon input rates to assess the fate of oil in Cook Inlet. The question of accumulation of crude oil components within the Inlet is considered from the above results and by direct analysis. Results indicate that hydrocarbon accumulation is less than our present limits of detection. Cook Inlet flushing is 90 per cent complete in 10 months. Experimental results show that unsupplemented Cook Inlet water effectively degrades Cook Inlet crude, that this biodegradation is essentially complete in the order of a few months, and that the biodegradation capacity of Cook Inlet is large. Thus biodegradation is more important than physical flushing in removing hydrocarbon pollutants from Cook Inlet. The methods and results are discussed in terms of their applicability to other areas.*

Cook Inlet (Figure 1) is a major estuary located in southcentral Alaska. The inlet itself is approximately 150 n miles long and 50 n miles wide at the mouth. The estuary is naturally divided north and south into two regions by the topographic features of the West and East Forelands. Two major extensions exist at the head of the inlet, Turnagain Arm which is 43 n miles long and Knik Arm which is 45 n miles long. The bottom of the inlet is extremely rugged and bottom sediments are sand and gravel. Mud flats exist along the north and west shore. The average depth of the inlet may be taken as 300 feet at the mouth, 120 feet near the forelands, and 60 feet near the head. Knik Arm averages 50 feet for about half its length and then rapidly shallows to a large mud flat.

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Turnagain Arm shallows within the first 10 n miles to large mud flats cut by many tidal channels.

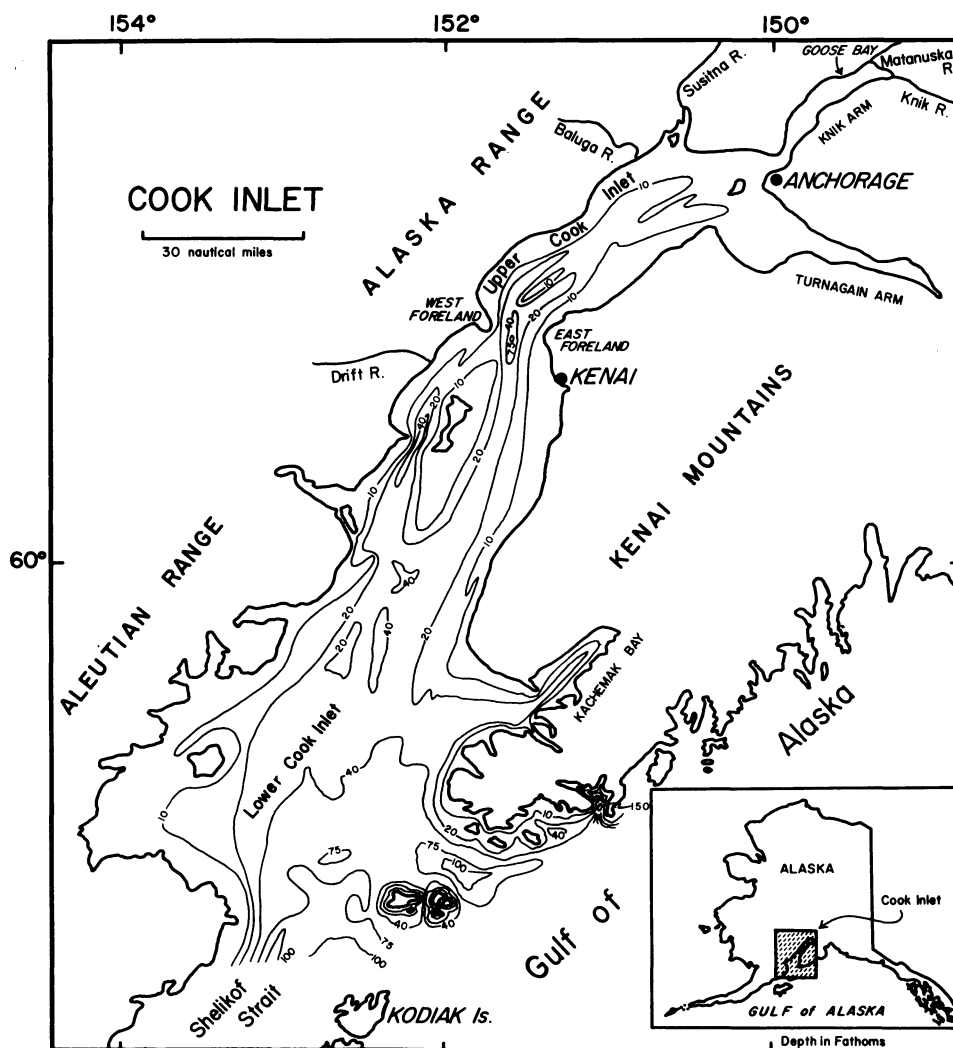
Tides and tidal currents are large.<sup>1</sup> The mean diurnal tidal range varies from 13.7 feet at the mouth to 29.6 feet at the City of Anchorage on Knik Arm. High velocity tidal currents are associated with these extreme tidal height differences. In the region of the forelands, velocities reach 3.8 knots and elsewhere, above these narrows, maximum velocities are in the range of 2.8 to 3.0 knots. In local areas velocities can be in excess of 5 knots. Large sheets of ice move back and forth with the tides during the three winter months.

Cook Inlet is a multiple-use estuary supporting large commercial fisheries, industrial and shipping usage, and extensive wildlife resources. Salmon, halibut, and crab are the more important commercialized fishery resources. Salmon and clams form the basis of recreational fishing. Additional wildlife resources include seal, beluga whale, ducks, geese, swan, and other waterfowl. Major oil industry related installations in Cook Inlet are shown in Figure 2. Offshore oil and gas wells are located from fifteen offshore platforms concentrated immediately north of the forelands. Underwater pipelines carry the oil to gathering facilities at East Forelands, Granite Point, and West Forelands where co-produced water is removed. From there, most of the oil goes to pipeline terminals for tanker shipment. Kenai Pipeline Terminal is located at East Forelands and the Cook Inlet Pipeline terminal is located at Drift River on the west shore.

Refineries and petrochemical plants are located at East Forelands and include the Standard Oil Refinery, Phillips Liquefied Natural Gas Plant, Collier Chemicals Ammonia

<sup>1</sup>Superior numbers refer to literature references at conclusion of text.

Figure 1.  
Map of  
Cook Inlet, Alaska.



Plant, and the recent Tesoro Refinery. The purpose of the present study was to quantitate Cook Inlet oil pollution in a manner that would furnish numbers to guide both industry and government. A secondary purpose was to develop both methods and results on the behavior and effects of petroleum in natural waters that would be of general applicability. Basic to this second point was the hope of a better method of monitoring pollution than the traditional, statistical inference of causes from difficulty measured biological damage. This latter approach is difficult in that continuous biological monitoring must be done wherever a pollution threat exists. More importantly, the questions needed to solve the pollution problem need to be answered with data on the specific mechanisms involved.

Three questions pertinent to oil pollution of natural waters need quantitative answers:

1. Fate of Oil:

Upon entering the water, where does the oil go, in what concentrations and compositions versus time and place?

2. Toxicity:

Given the information of 1. what are the specific physical or biochemical toxic mechanisms of damage?

3. Treatment of Spills:

Based upon firm knowledge of 1. and 2., how

should a major spill be treated to minimize damage?

Progress must of necessity be measured in relation to these vigorous goals of ultimate solution to oil pollution. The bulk of the present work is addressed to the first question, the beginning, but the results have implications for topics 2. and 3.

Consider Cook Inlet to be an open system 150 n miles long, 50 n miles wide at the mouth, with a total volume of  $1.7 \times (10)^{13}$  ft.<sup>3</sup> Consider the fate of oil question initially as an oil material balance about the Cook Inlet system. The rate of oil accumulation must equal the rate of input minus the rate of output and minus the rate of consumption thus:

$$\text{Accumulation} = \text{Input} - \text{Output} - \text{Consumption}$$

We have tried to elucidate all the mechanisms affecting each term of the above equation and tried to measure their corresponding rates.

### Results

**Input.** The input term can be considered to be the unknown for which the equation is to be solved. However, an estimate by independent means is most helpful at this time as a check on the magnitude of the other terms. To estimate the order of magnitude of the accidental input term, data available was obtained from Federal and

State sources for years 1966, 1967, 1968, and 1969 on oil sightings in Cook Inlet gathered on:

1. random flights, and
2. flights following reports by industry and others.<sup>2</sup>

These data were combined with data on the amount of oil per unit areas of slicks (100 gallons/miles,<sup>2</sup> 3 hours old; and 30 gallons/miles,<sup>2</sup> 8 hours old), from measurements made on a large pipeline incident, and from a small experimental spill. Work was done from a Dehaviland Beaver float plane. In the case of the experimental spill, 20 gallons of Cook Inlet crude were poured on the water in the region of the forelands and were followed for a day by measuring and sampling from the air and on the water. The results of combining the above data indicate an input term of 2,000 - 10,000 bbl/year, almost all of it in the form of large, reported accidental incidences. The contribution from random sightings on a yearly basis ranged from 20-300 bbl/yr. The waste water from routine process streams discharged into the inlet from platforms, terminals, gathering facilities, and refineries and petrochemical plants contribute about 7,500 bbl/yr. of oil. This total input of 9,500 - 17,500 bbl/yr. corresponds to approximately 0.03 per cent of the total oil produced or handled in Cook Inlet.

*Output.* One important output term is due to the physical flushing of the estuary. The Cook Inlet estuary can be classified<sup>3</sup> according to physical circulation deduced from salinity distribution. Surface salinity and temperature distributions (Figures 4 and 5) along with detailed depth profiles presented elsewhere,<sup>4</sup> show that Cook Inlet can be typed as a two-layer flow estuary with intense vertical mixing which approaches a vertically homogeneous estuary with lateral variation due to coriolis force in the region north of Kalgin Island. An overall analysis of mixing in an estuary, in order to estimate the rate of removal of a pollutant introduced into it, is usually done by using fresh water as a tracer. Flushing of an estuary depends on the river input rate and salt water replacement rate due to mixing entrainment. A simple salt balance yields the following expressions assuming steady state:<sup>3,5</sup>

$$R(O) + V_i(S_o) = (R + V_i) S_a \quad (1)$$

or

$$V_i = \frac{RS_a}{(S_o - S_a)} \quad (2)$$

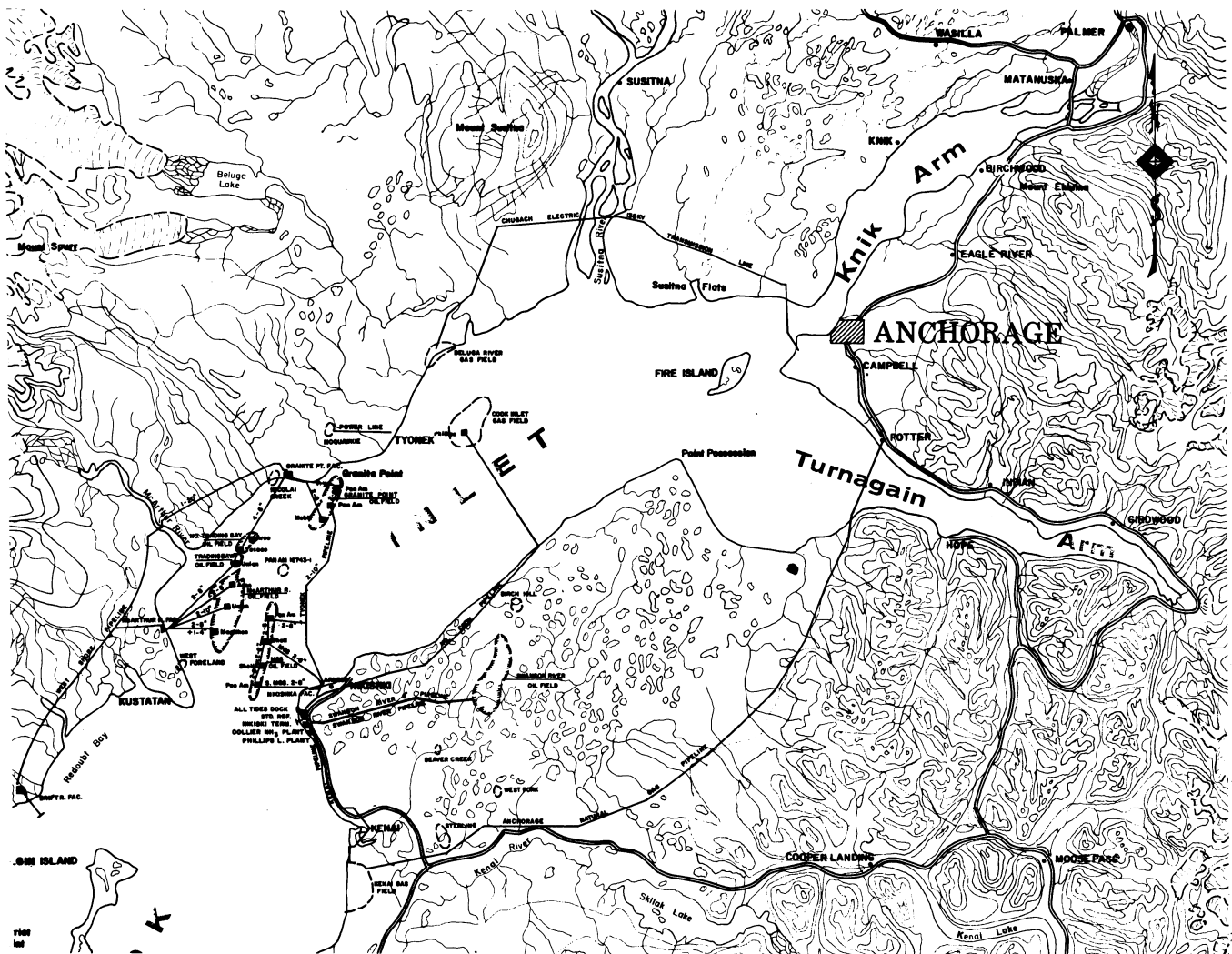
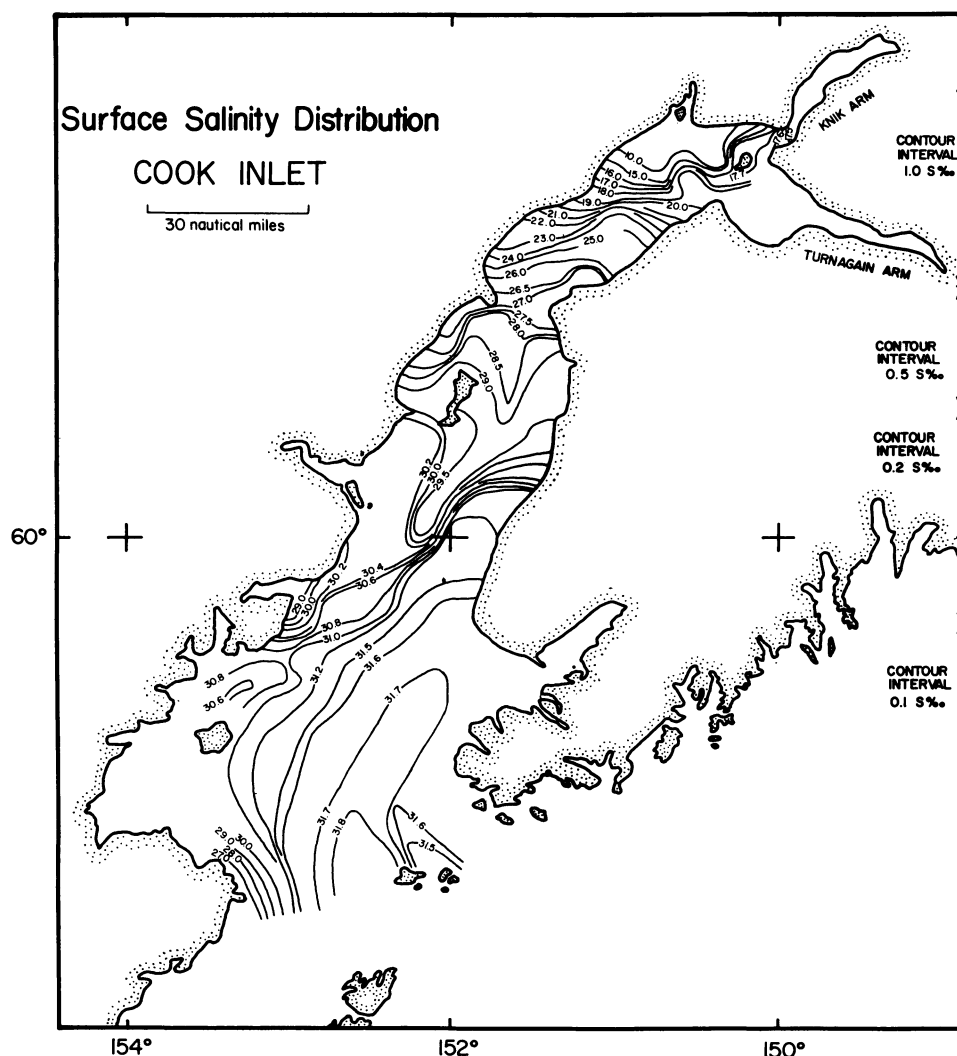


Figure 2. Major oil industry related installations, Cook Inlet, May 1968.

Figure 3.  
Surface salinity distribution,  
Cook Inlet, May 1968.



Where  $R$  is the fresh water flow rate into the estuary,  $S_o$  is the salinity of the saline source water at the mouth (at depth),  $V_i$  is the rate of salt water inflow due to entrainment, and  $S_a$  is the average salinity of the estuary. Using June salinity and river flow data<sup>5</sup> values of  $S_o=32.0$  per cent,  $S_a=29.7$  per cent and  $R=1.02(10)^5$  ft.<sup>3</sup>/sec. were obtained. Using these values in the salt balance equation, the value of  $V_i=1.32(10)^6$  ft.<sup>3</sup>/sec. is obtained which is equivalent to  $4.40(10)^{13}$  ft.<sup>3</sup>/yr for the total input term  $(R+V_i)=F$ . Winter time river flow rates are only 5 per cent of this June value which would vastly decrease the flushing flow if the salinity distribution were constant. This reduced flushing would be effective for approximately half the year. However, Bowden<sup>3</sup> states that flushing times are not a strong function of seasonal river input variations based upon seasonal analysis of other estuaries. Since in Cook Inlet, tidal turbulence, independent of season, is a major factor in salt transport up the estuary, the  $V_i$  term would be expected to remain high. The very limited winter salinity data available also indicate a significant increase as would be expected. Therefore, using the above figures the approximate time for an introduced pollutant to be reduced to 10 per cent of its original concentration by flushing is given by the following expression with  $V$  being the total inlet volume:

$$t_{90\%} = \frac{(V)}{(R + V_i)} \ln 10 = 10 \text{ months} \quad (3)$$

Similarly an oil material balance

$$V \frac{dc}{dt} = I - Fc \quad (4)$$

Where  $I$  is the yearly input of crude oil and  $c$  is the resulting crude oil concentration in the water, results in a steady state crude concentration of 0.82 to 1.5  $\mu\text{g}/\text{l}$  based upon yearly inputs of 9,500 to 17,500 bbl/yr. (1  $\mu\text{g}/\text{l}$  = 1ppb) Time to reach 90 per cent of the steady state value would be approximately one year, starting from a zero concentration.

Output due to evaporation was determined by field measurements because of the difficulty in simulating air-sea interaction and interfacial emulsion properties in the laboratory. Two sets of measurements were performed with similar results. One involved sampling oil surfacing directly above the Santa Barbara leak and comparing the gas chromatographic spectrum with samples removed from the spill. The second involved sampling a small experimental spill of Cook Inlet crude in Cook Inlet over a period of one day by use of a float plane. Average wind speed throughout the day was 9-12 knots for this experimental spill.

Results of the latter experiment, shown in Figure 5, show that compounds below about  $C_{12}$  are significantly reduced in 8 hours. This reduction was presumably chiefly due to evaporation, although solubilization of the lower molecular compounds in the water phase also occurs. From these results, the major fate of gasoline and kerosene fractions is probably evaporation to the atmosphere, with gasoline being essentially gone in 8 hours. For light refined products or for crude oils with significant fractions in this boiling point region (e.g. Cook Inlet crude), this mechanism can thus be important in significantly reducing the water pollution.

**Consumption.** The major petroleum consumption term is microbial oxidation to  $CO_2$  and  $H_2O$ . Field and laboratory observations were made in order to understand the mechanisms and rates involved. Repeated aerial observations of both small experimental spills and large accidental spills indicated the apparent half life of a surface crude oil slick in Cook Inlet was less than a day. Even in the case of a 1,000 bbl. pipeline break, the oil had completely disappeared from the surface in 3-4 days. Laboratory results show that the microflora excrete effective emulsifying agents which help the process considerably. Of course the great turbulence of Cook Inlet, driven by 20-30 foot tides and up to 5 knot currents, is a major factor.

Microorganisms capable of oxidizing crude oil can be found throughout Cook Inlet. Plate counts and most probable number determinations show populations of approximately  $10^4$  organism/liter with approximately 10 per cent of this population capable of utilizing crude oil. Of interest is the fact that a significant number of oil oxidizing microorganisms enter into the oil phase, rather than remaining in the aqueous phase and working on the surface of oil droplets. Particles of water in oil emulsion, mixed down into the water phase are literally micro-environments, being oxidized by microorganisms existing in both phases. These particles are more dense than sea water due to the presence of oxidized cell material and biomass and oxidized hydrocarbon intermediates. However, moderate turbulence keeps these particles in suspension.

Several microorganisms were isolated and found to use kerosene as the sole source of carbon. Chromatographic analysis indicated that various species selected different components from the hydrocarbons for their carbon and energy sources. Maximum growth rates ranged from 0.02 g cells/ g cells/ hr. in a mineral salts kerosene medium to 0.35 hr.<sup>-1</sup> on a complete medium.<sup>4</sup> These rates are reduced by a factor of 7 at 5°C, the prevailing summer temperature of Cook Inlet (Figure 4).

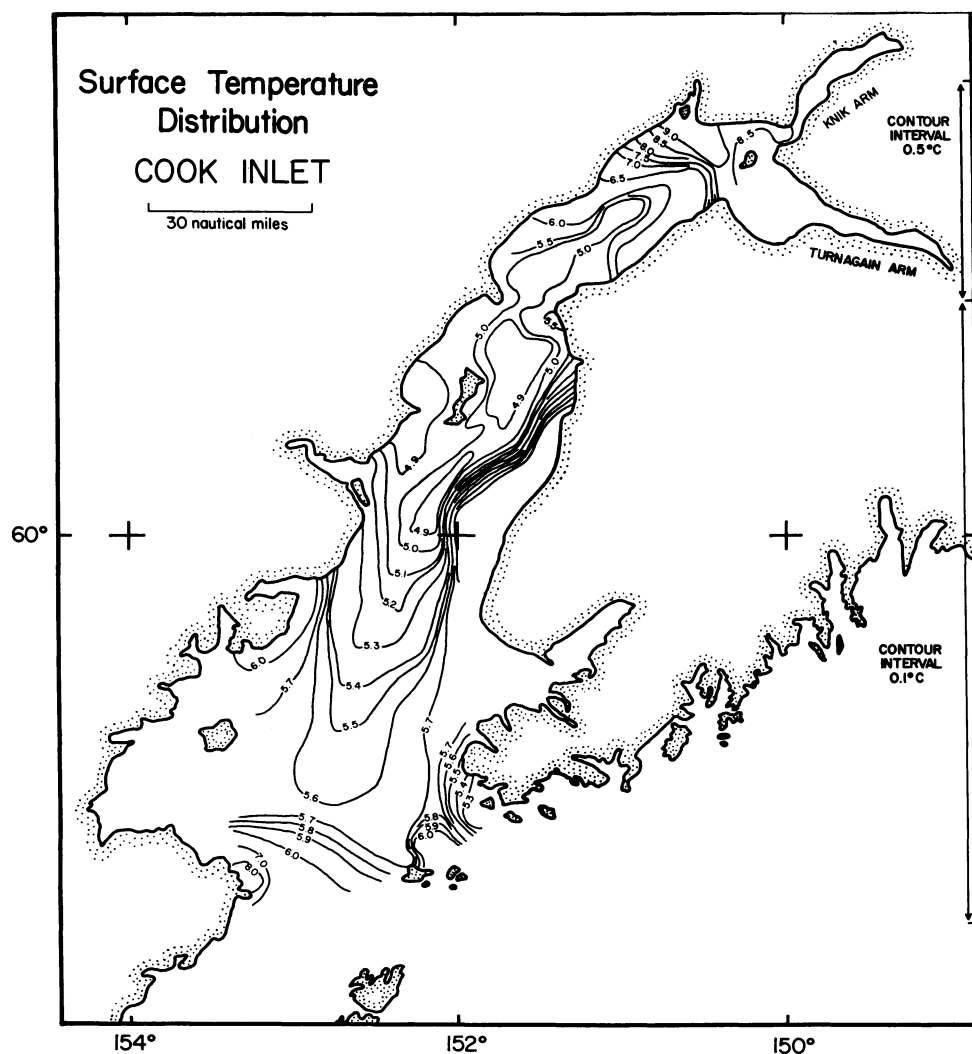


Figure 4.  
Surface temperature  
distribution, Cook Inlet,  
May 1968.

For dilute populations in the sea, the supply of growth factors is probably adequate, particularly at low temperatures where growth rates are low with respect to diffusion transport. Nitrogen and phosphorous are in excess so that, barring increased inhibitor effectiveness at these low populations by species such as heavy metals, growth rates probably approach those in a complete medium. At this rate a 1000 fold population increase would require 5.7 days and complete oxidation of a 1 mg droplet of biodegradable oil would require 17 days after inoculation by a single bacterium. Populations in sterile bottles filled with Cook Inlet water and supplemented with autoclaved Cook Inlet crude oil were  $10^9$  per liter after 5 days at  $10^\circ\text{C}$  which is in agreement with the above growth rate and initial populations, assuming the success of collisions between the initial population and the oil was extensive during the first day.

Degradation of the heavy crude is probably slower than that of the fraction below  $\text{C}_{25}$ . Rather heavy artificial slicks confined in a carboy were rich in normal hydrocarbons above this size after two months' incubation with a mixed culture. Degradation of lesser concentrations of crude, 20 mg/l, appeared to be essentially complete after two months at  $10^\circ\text{C}$  and less than 5 per cent could be recovered at the end of one year. Artificial slicks on sea water surfaces in stirred carboys were clearly different in inoculated and sterile carboys at the end of three days, and degrading slicks broke up at the end of ten. The presence of silt appeared to have no effect on the process but mixing was extremely important. Slicks in completely unmixed carboys were very stable even when inoculated. Thus it appears that naturally occurring slicks are rapidly inoculated and broken up during the first day after a spill in the very turbulent Cook Inlet. Biodegradation then proceeds first in the micro-environments of the water in oil emulsion, later in the oil drops, and is probably essentially complete in one to two months and certainly before surrounding water is flushed out of the inlet.

Ultraviolet oxidation of crude oil is estimated to be negligible compared to biodegradation and flushing because of the low radiant energy input and the short half life of a thin surface film of oil. However, few data exist on this mechanism.

**Accumulation.** Independent estimates of the accumulation term were attempted by direct chemical analysis for hydrocarbons in the existing water and sediments of Cook Inlet. Experimental work is also in progress on two other independent methods using  $^{14}\text{C}$  radiocarbon<sup>6</sup> and  $^{12}\text{C}$   $^{13}\text{C}$  stable carbon isotope<sup>7</sup> analysis of dissolved and particulate organic matter from Cook Inlet. A carbon recovery apparatus<sup>8</sup> based on ultraviolet oxidation of the organic matter in sea water is in use for the  $^{14}\text{C}$  measurements. Natural carbon in Cook Inlet and Cook Inlet crude seem to have nicely separated  $^{12}\text{C}/^{13}\text{C}$  ratios. We do not at present have enough data to make conclusions from these latter two experimental results.

Direct chemical analysis for hydrocarbons from water and sediments was by gas chromatography preceded by an extractive concentration step. A considerable body of knowledge on the origin of petroleum and the migration in reservoirs<sup>10-20</sup> shows recent sediment  $\text{C}_{20}\text{-C}_{33}$  n-alkane concentrations to be 10-70  $\mu\text{g/g}$  dry weight. One

reference<sup>21</sup> for surface waters shows concentrations from 0.2 to 3.8  $\mu\text{g/l}$  and two sea water samples at less than 0.2  $\mu\text{g/l}$  total  $\text{C}_{20}\text{-C}_{33}$  n-alkanes. Blumer<sup>9</sup> has identified by gas chromatography, the presence of  $\text{C}_{14}$  to  $\text{C}_{33}$  straight chain hydrocarbons, branched paraffins at an order of magnitude less concentration, isoprenoid hydrocarbons, and at least two olefinic hydrocarbons (squalene and a polyunsaturated derivative of heneicosane). No concentrations or spectra are given. Thus the knowledge of natural hydrocarbons in aqueous systems is very limited; however, it is known that they are present and that their concentrations are extremely low.

In the present work, four liters of water, sampled with a large volume sampler of glass, stainless, and teflon, was extracted three times with pre-purified n-pentane and the extracts combined to 500 ml. Evaporation at  $36^\circ\text{C}$  with a small reflux condenser reduced the volume to approximately 1 ml. Silica gel chromatography<sup>9</sup> was used to isolate saturated hydrocarbons and the isolate in pentane was evaporated to a fraction of a ml and injected into a temperature programmed gas chromatograph equipped with a flame ionization detector. A fifty foot 0.025 inch diameter open tubular capillary column with 5 per cent OV-17 on the wall coating support was used. Detection limits as determined by internal standards was of the order of 0.02  $\mu\text{gm/l}$  for individual hydrocarbons in the  $\text{C}_{10}\text{-C}_{25}$  molecular weight range or about 20  $\mu\text{g/l}$  of Cook Inlet crude. Analysis of samples of water and suspended sediments from 70 stations distributed throughout the Inlet showed completed negative results to the detection limits quoted above. Thus these results indicate that the Cook Inlet system is not grossly polluted by hydrocarbons. However, the present detection limits do not allow an independent estimate of the relative effectiveness of flushing and biodegradation. Work is continuing on improving these detection limits by one or two orders of magnitude by eliminating the sample splitter for the open tubular column or by use of conventional columns.<sup>21</sup>

Data on light gas analysis ( $\text{C}_1\text{-C}_5$ ) showed indicated possible minute leakage from the reservoir structures; however, the amounts involved are infinitesimal in terms of hydrocarbon pollution. Accumulation of microbiologically fractionated refractory compounds appeared to be negligible and is discussed in C above and elsewhere.<sup>4</sup>

The possible buildup in Cook Inlet of toxic transition metals from crude oil pollution was considered. If the highest value from above for the crude input is assumed and combined with the general ppm concentrations of trace metals in crude, then the steady state level of trace metal concentrations due to oil input is limited by physical flushing to a value three orders of magnitude below natural levels. Thus this danger does not seem to be a real one in Cook Inlet. Also, our biodegradation rates run with Cook Inlet crude, would consider localized concentrations of trace metals effecting these rates.

## Discussion

Major effects of accidental spills on the environment of Cook Inlet are not clear. Certainly spills would have little effect on the water chemistry such as carbon, oxygen and nitrogen which are usual indicators of pollution. The more important effects of a large spill might be reduction of the

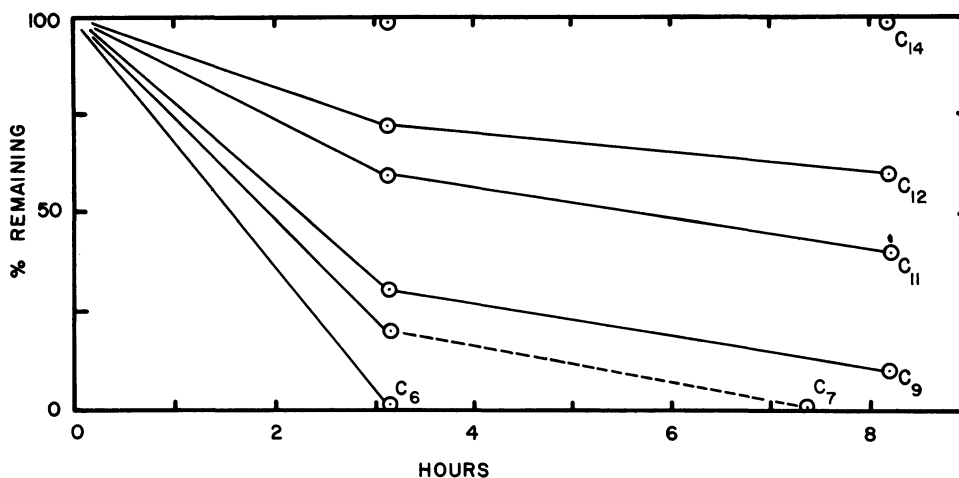


Figure 5.  
Evaporative loss of low  
molecular weight hydrocarbons  
in an experimental spill  
of Cook Inlet crude  
in the forelands regions  
of Cook Inlet.

aesthetic quality of beaches, fouling of fishermen's gear and oiling of water fowl. Continuous oil slicks on the surface are probably of more concern than similar quantities of oil dispersed below the surface. Only at the surface does oil contribute to the aforementioned problems.

Aside from prevention, no technology exists to substantially minimize damage from a large accident. Recognition of biodegradation as the fastest process in removal of hydrocarbons from Cook Inlet has implications for the treatment of oil spills. For example, detergents might be designed to act in concert with this process.

Given potentially high rates and capacity of the biodegradation mechanism methods of sinking oil spills into the water column may well be practically, as well as ecologically, attractive. The large quantities of Cook Inlet sediment available might be investigated as a type of vehicle, after proper treatment, to do this.

Some of the information gathered here is applicable elsewhere. For example, biodegradation appears to be a significant low temperature process. Lack of mixing is a significant hindrance to this mechanism. Hence, a potential oil spill problem area, such as the Arctic, should be investigated in terms of the physical stability of oil in pack ice and in the comparatively poorly mixed surface waters. The biodegradation mechanism should be assessed under these conditions.

## Conclusions

1. Estimated input of petroleum hydrocarbons from accidental spills and effluents is approximately 10,000 to 17,000 barrels per year or approximately 0.03 per cent of the total produced and handled in Cook Inlet.
2. The half life of an oil slick on the surface of Cook Inlet is less than a day.
3. Tide driven turbulence is the chief factor in breaking up oil slicks on the surface of Cook Inlet.
4. Generally, strong tidal currents and their geometry tend to keep oil which has been spilled out in the inlet away from beaches, with the exception of Kalgin Island.
5. A microflora of hydrocarbon oxidizing organisms exists at a concentration of about  $10^3/1$  which functions as an inoculum and suggests the present of transient hydrocarbons.

6. Biodegradation of Cook Inlet crude oil in Cook Inlet is essentially complete in one to two months.
7. Tidal and river driven flushing reduces Cook Inlet components within the inlet by 90 per cent in about 10 months.
8. Evaporation effectively removes components smaller than about C<sub>12</sub> within 8 hours.
9. Accumulated petroleum hydrocarbons in the range C<sub>10</sub>-C<sub>25</sub> were below the limits of detection, 0.02  $\mu\text{g}/1$ , in Cook Inlet waters and suspended sediments. Trace transition metals from crude oil were found to be limited by physical flushing to levels three orders of magnitude below natural levels.

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